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A SEARCH FOR ^{70}Zn ANOMALIES IN METEORITES

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ABSTRACT

No ^{70}Zn isotopic anomalies have been detected in primitive meteorites to a level of precision of less than 40 parts per million (2σ). Any pre-existing nucleosynthetic anomaly on ^{70}Zn was averaged out by mixing in the solar nebula before planetary accretion in the solar system. Because neutron-rich nuclides ^{70}Zn and ^{60}Fe are produced by similar nucleosynthetic processes in core-collapse supernovae, the homogeneity of ^{70}Zn in meteorites limits the possible heterogeneity of extinct ^{60}Fe radioactivity in the early solar system. Assuming that Fe and Zn have not been decoupled during incorporation into the solar system, the homogeneity of the ^{70}Zn/^{64}Zn ratio measured here implies that the ^{60}Fe/^{56}Fe ratio was homogenized to less than 15% dispersion before the formation of planetary bodies. The lack (Zn, Ni, Fe) or presence (Ti, Cr) of neutron-rich isotopic anomalies in the iron mass region may be controlled by the volatility of presolar carriers in the nebula.

Key words: astrochemistry – minor planets, asteroids – nuclear reactions, nucleosynthesis, abundances – planetary systems: protoplanetary disks – solar system: formation

1. INTRODUCTION

Since the 1970s, various isotopic anomalies have been observed at the mineral scale for many elements in certain types of Ca–Al-rich inclusions (CAIs) from the Allende chondrite named FUN (Fractionated and Unknown Nuclear effects) inclusions (e.g., Wasserburg et al. 1977; Clayton 1978; McCulloch & Wasserburg 1978a, 1978b). These mineral scale anomalies are usually ascribed to incomplete mixing of the products of stellar nucleosynthesis in the early solar system (Wasserburg et al. 1979), but chemical origins have also been proposed (Fujii et al. 2006; Robert 2004). More recently, improvements in analytical methods and instrumentation have permitted resolution of progressively smaller isotopic anomalies for several elements (Ca, Cr, Ti, Ni, Mo, Ru, Ba, Sm, Nd, W) in bulk meteorites (Podosek et al. 1997; Dauphas et al. 2002; Yin et al. 2002; Chen et al. 2003; Hidaka et al. 2003; Dauphas et al. 2004; Papanastassiou et al. 2004; Andreasen & Sharma 2006; Ranen & Jacobsen 2006; Carlson et al. 2007; Leya et al. 2008; Qin et al. 2008; Simon et al. 2009; Trinquier et al. 2007, 2009).

Measuring the abundance of several isotopes produced by the same process can provide useful constraints on the origin of isotopic anomalies. Neutron-rich nuclides such as ^{58}Fe, ^{60}Fe, and ^{64}Ni were synthesized in core-collapse supernovae or asymptotic giant branch (AGB) stars, by neutron capture reactions (Clayton 2003). They were not produced inside the solar system by irradiation. Thus, their potential nucleosynthetic anomalies should be correlated. Dauphas et al. (2008) measured the isotopic abundances of ^{58}Fe together with ^{64}Ni and ^{60}Ni (produced by the decay of ^{60}Fe, $t_{1/2} = 1.49$ Myr) to quantify the possible heterogeneity in the distribution of ^{60}Fe at the scale of bulk meteorites in the early solar system. Iron-58 and ^{64}Ni did not show any departure from terrestrial composition at precisions of ∼0.3–0.5 ϵ (deviation in part per 10,000 relative to the composition of a terrestrial reference material). These results, together with the absence of fossil ^{60}Fe in the same samples (at the whole-rock scale), argue in favor of the injection of ^{60}Fe in the early solar system and its homogenization before the formation of planetary bodies, in agreement with dynamic modeling (Boss 2007).

In this Letter, we extend that study to ^{70}Zn, which is another neutron-rich nuclide from the Fe peak, synthesized by processes also responsible for the formation of ^{60}Fe in cc-SN (Figure 1). Zinc has four stable isotopes: ^{64}Zn (48.63%), ^{66}Zn (27.90%), ^{67}Zn (4.10%), and ^{68}Zn (18.75%). Zinc-70 (0.62%) is a radioactive isotope with a half-life (5×10^{14} yr) sufficiently long in comparison to the age of the solar system that it can be considered stable. Zinc-70 isotopic anomalies have already been detected in two FUN inclusions, C1 and EK1-4-1, by thermal ionization mass spectrometry (TIMS), while no anomalies were reported in other CAIs or whole-rock samples with a precision $> 2\ \epsilon$ (Gawinowski et al. 1989; Loss & Lugmair 1989; Völkening & Papanastassiou 1990). Recent studies conducted using a multi-collector inductively coupled plasma mass-spectrometer (MC-ICP-MS) showed that the four most abundant isotopes of Zn are homogeneously distributed (Luck et al. 2005; Moynier et al. 2007) at a precision of ∼0.5 ϵ (2σ); however, these studies were mostly focused on mass-dependent isotopic fractionation and did not report the very low abundance and analytically challenging ^{70}Zn. For this study, we developed an original method for measuring ^{70}Zn/^{64}Zn ratios at high precision using a MC-ICP-MS.

2. SAMPLES AND ANALYTICAL PROCEDURES

We analyzed the Zn isotopic compositions of whole-rock samples of Ornans (CO3), Allende (CV3), Murchison (CM2), Forest Vale (H4), and Indarch (EH4). For each sample, ∼500 mg were dissolved under pressure in Paar bombs in a mixture of HNO_3/HF. A subsequent step in aqua regia was carried out to ensure the dissolution of refractory phases. The protocol for chemical purification was guided by the necessity of minimizing isobaric interferences with Zn isotopes, particularly from ^{64}Ni on mass 64 and ^{70}Ge on mass 70. Zinc was purified by anion-exchange chromatography using HBr/HNO_3, following the protocol of Moynier et al. (2006, 2009).

Table 1
Zn Isotopic Composition of Bulk Meteorites (see the text for details of the ϵ notation)

Sample Name	Type	ϵ^{64}Zn	ϵ^{66}Zn	ϵ^{67}Zn	ϵ^{68}Zn	ϵ^{70}Zn	n^{a}
Forest Vale	H4	0	−0.18 ± 0.29	0.42 ± 0.19	0	0.32 ± 1.07	10
Allende	CV3	0	0.16 ± 0.30	−0.05 ± 0.10	0	−0.33 ± 0.73	20
Murchison	CM2	0	0.07 ± 0.27	0.00 ± 0.17	0	0.14 ± 0.72	21
Ornans	CO3	0	0.72 ± 0.86	−0.01 ± 0.26	0	0.27 ± 1.12	9
Indarch	EH4	0	0.06 ± 0.31	−0.04 ± 0.27	0	−0.47 ± 1.23	6
Terrestrial standard	···	0	0.01 ± 0.07	0.00 ± 0.04	0	−0.03 ± 0.12	28
Chondrite weighted average	···	0	0.04 ± 0.14	0.03 ± 0.07	0	−0.03 ± 0.40	···

Notes. ^{x}Zn/^{64}Zn normalized to ^{68}Zn/^{64}Zn = 0.568828, where x = 66, 67, 68, and 70.
[a] Number of standard-sample bracketing measurements.

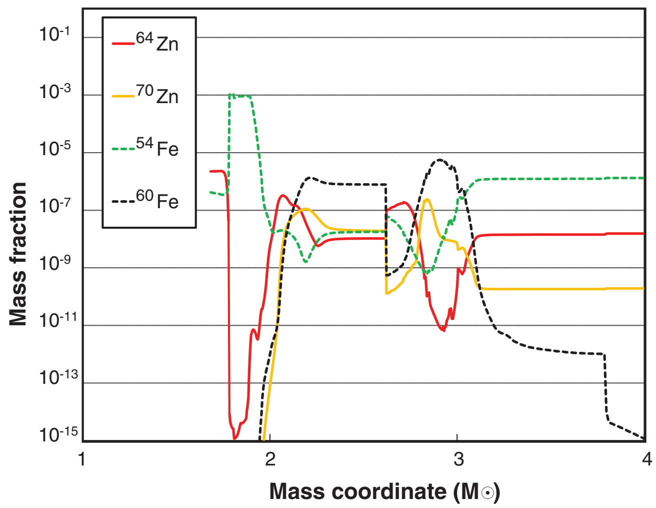

Figure 1. Post-supernova profile of ^{54}Fe, ^{60}Fe, ^{64}Zn, and ^{70}Zn abundances as a function of mass coordinate for a 15 $M_{\odot}$ supernova Type II progenitor computed from zonal yields 25,000 s after core bounce (see Rauscher et al. (2002) for details). While ^{54}Fe and ^{64}Zn are produced as radioactive progenitors in the internal regions of the supernova by nuclear statistical equilibrium, the neutron-rich isotopes ^{60}Fe and ^{70}Zn are produced in more external regions by neutron-capture reactions on pre-existing Fe and Zn isotopes.

Zinc isotopic compositions were measured on a Thermo Scientific Neptune MC-ICP-MS at the Origins Laboratory of the University of Chicago. The purified Zn solutions (1 ppm in 0.3 M HNO_3) were introduced into the MC-ICP-MS using an Apex-Q+Spiro inlet system operated with Ar alone and a 100 μL minute^{-1} PFA nebulizer. On-peak zero intensities from a blank solution were subtracted from all measurements. Because the ion beam array did not allow the acquisition of ^{62}Ni (used for the correction of isobaric interferences on ^{64}Zn) and ^{70}Zn simultaneously, a dynamic mode of two magnetic field settings was applied. The intensities on masses 64, 66, 67, 68, and 70 were measured in the first field setting and the intensities on the masses 62, 64, and 66 were measured in the second field setting. All collectors but L4 (^{64}Zn) were connected to 10^{11} Ω amplifiers. To get a high signal on the minor isotope ^{70}Zn and avoid saturation on the most abundant isotope, ^{64}Zn, a 10^{10} Ω amplifier was connected to L4. The resulting ion beam intensity was ∼1 V on ^{70}Zn.

3. RESULTS

Isotope ratios are reported in Table 1 in ϵ units (deviation in part per 10,000 relative to the composition of the reference material JMC-Lyon 3-0749L; Equation (1)) after internal normalization to a ^{68}Zn /^{64}Zn ratio of 0.568828 using the exponential law (Russell et al. 1978; Maréchal et al. 1999),

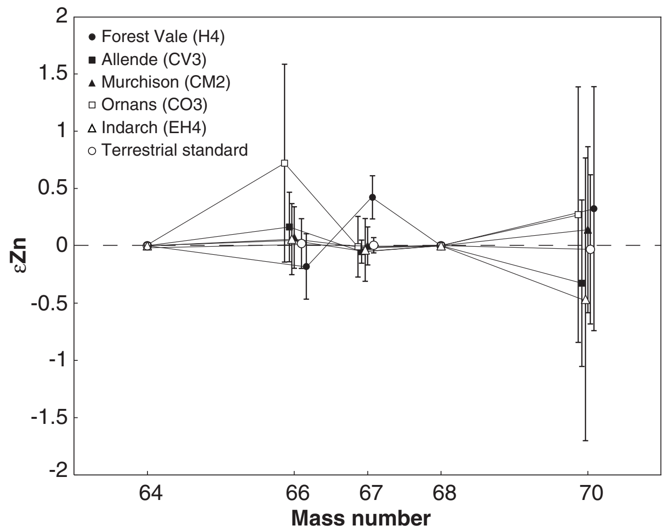

Figure 2. Zn isotopic data in chondrites. No ^{70}Zn anomalies were detected. The average ϵ^{70}Zn of the five meteorites is −0.03 ± 0.40 ϵ.

$$\epsilon^{x}\mathrm{Zn} = \left(\frac{(^{x}\mathrm{Zn}/^{64}\mathrm{Zn})_{\mathrm{sample}}}{(^{x}\mathrm{Zn}/^{64}\mathrm{Zn})_{\mathrm{JMC\text{-}Lyon\ 3\text{-}0749L}}} - 1 \right) \times 10{,}000, \quad (1)$$

x = 66, 67, or 70.

As shown in Figure 2, all meteorites analyzed in this study have isotopic compositions identical to terrestrial. Forest Vale shows a small ^{67}Zn excess (+0.42 ± 0.19 ϵ), which may represent normal statistical fluctuation and will not be discussed hereafter. The average of ϵ^{70}Zn among the five chondrites is −0.03 ± 0.40 (95% confidence interval of the weighted average). The absence of ^{70}Zn anomalies at the whole-rock scale in meteorites agrees with previous studies (Gawinowski et al. 1989; Völkening & Papanastassiou 1990) but with significantly improved precision (by a factor of 2–3; Figure 3).

4. DISCUSSION

These results imply that at the scale of chondrite parent bodies the ^{70}Zn/^{64}Zn ratio was homogenously distributed in the early solar system and that any potential pre-existing nucleosynthetic anomalies were averaged out by mixing in the solar nebula by the time of planetary body accretion. This observation is consistent with similar conclusions reached for other moderately volatile elements from the Fe peak (Cook et al. 2006; Quitté et al. 2006; Moynier et al. 2007; Dauphas et al. 2008; Chen et al. 2009). On

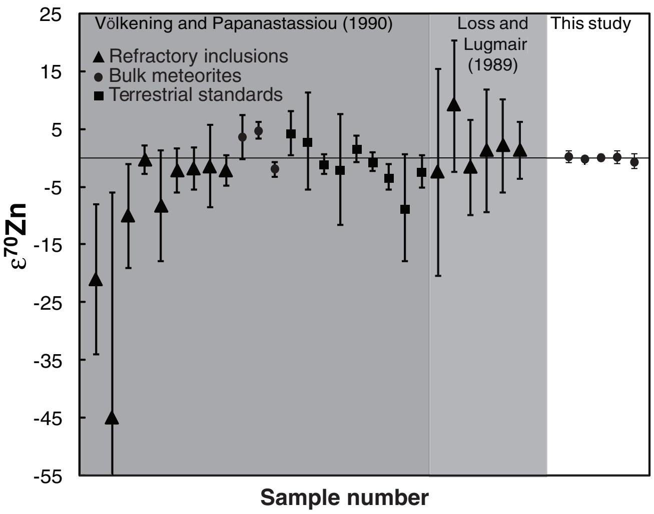

Figure 3. Comparison between ^{70}Zn data from this study and Loss & Lugmair (1989) and Völkening & Papanastassiou (1990).

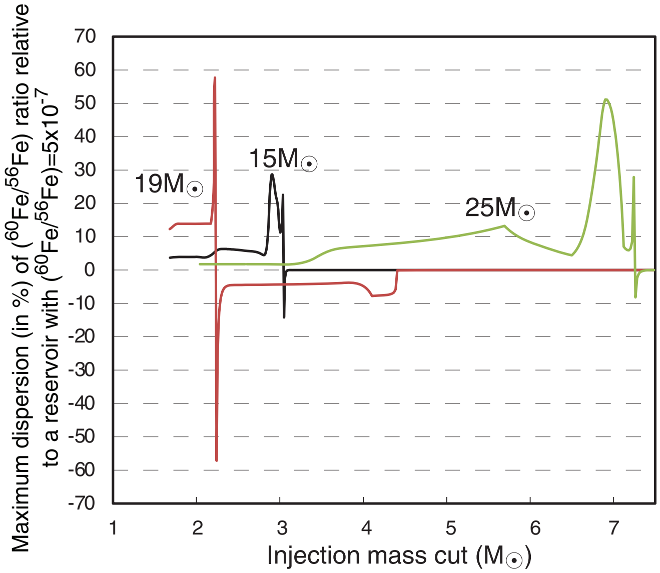

Figure 4. Maximum dispersion of ^{60}Fe/^{56}Fe ratio (in %) relative to a reservoir with ^{60}Fe/^{56}Fe $= 5 \times 10^{-7}$ as a function of injection mass cuts of cc-SN (see the text for details). The yields for 15, 19, and 25 $M_\odot$ cc-SN progenitors from Rauscher et al. (2002) were used in Equation (2) to calculate the curves in this figure.

the other hand, Cr and Ti show clear isotopic anomalies at the meteorite parent-body scale (Niederer 1985; Niemeyer 1988; Shukolyukov & Lugmair 2002; Leya et al. 2008; Qin et al. 2008; Trinquier et al. 2009). This may be related to the presence of refractory presolar carriers of isotopic anomalies for Ti and Cr in meteorites that have so far eluded identification.

An important implication of this study is that the distribution of ^{70}Zn can be used to probe the homogeneity of the short-lived nuclide ^{60}Fe. The isotopes of Fe (and Zn) have been produced by different nucleosynthetic processes (nuclear statistical equilibrium and neutron capture) in different stellar environments (e.g., AGB stars, supernovae). The Fe (and Zn) isotopic composition of the solar system is the sum of this production since the formation of the Galaxy. Therefore, the Fe (and Zn) isotopic compositions of the individual star(s) which made ^{60}Fe found in meteorites would be expected to have exotic composition(s) relative to average solar system material. Thus, if some part of the solar system had deficits or excesses of ^{60}Fe, correlating effects in ^{70}Zn are expected.

If Fe and Zn had not been decoupled during the injection, the collateral effect on ^{60}Fe corresponding to heterogeneous distribution of ^{70}Zn can be calculated by adapting Equation (3) from Dauphas et al. (2008) to ^{70}Zn:

$$\Delta^{60}\mathrm{Fe}/^{56}\mathrm{Fe} = \frac{1}{c}\frac{1+\rho^{56}_{\mathrm{Fe}}}{\rho^{70}_{\mathrm{Zn}} - \mu^{70}_{\mathrm{Zn}}\rho^{68}_{\mathrm{Zn}}}\frac{\left(^{60}\mathrm{Fe}/^{56}\mathrm{Fe}\right)_{\text{cc-SN/AGB}}}{e^{\lambda_{60}\Delta t}\times 10^4} \times \Delta\epsilon^{70}\mathrm{Zn}, \tag{2}$$

where Δ^{60}Fe/^{56}Fe is the dispersion of the ^{60}Fe/^{56}Fe ratio of nebular reservoirs characterized by chondritic Fe/Ni ratio relative to a reference reservoir with a ^{60}Fe/^{56}Fe value of 5×10^{-7}, corresponding to what is inferred in the chondrule-forming region at the time of condensation of the first solids in the nebula (Tachibana et al. 2006). $\Delta\epsilon^{70}$Zn is the maximum variability of ϵ^{70}Zn in the early solar system estimated from our measurements to be equal to 0.40. $\rho^{70}_{\mathrm{Zn}} = (^{70}\mathrm{Zn}/^{64}\mathrm{Zn})_{\text{cc-SN/AGB}}/(^{70}\mathrm{Zn}/^{64}\mathrm{Zn})_{\mathrm{CHUR}} - 1$; $\mu^{70}_{\mathrm{Zn}} = (70-64)/(68-64)$; $c = (^{64}\mathrm{Zn}/^{54}\mathrm{Fe})_{\text{cc-SN/AGB}}/(^{64}\mathrm{Zn}/^{54}\mathrm{Fe})_{\mathrm{CHUR}}$, λ_{60} is the decay constant of ^{60}Fe, $\rho^{56}_{\mathrm{Fe}} = (^{56}\mathrm{Fe}/^{54}\mathrm{Fe})_{\text{cc-SN/AGB}}/(^{56}\mathrm{Fe}/^{54}\mathrm{Fe})_{\mathrm{CHUR}} - 1$, and Δt is the decay interval between production of ^{60}Fe in cc-SN or AGB stars and its injection in the solar system. For simplicity, injection from only one star is considered while several massive stars could have contributed to the synthesis of ^{60}Fe found in meteorites (Gounelle et al. 2009). Expected isotopic anomalies were computed using the yields for 15, 19, and 25 $M_\odot$ Type II supernovae progenitor (data from Rauscher et al. 2002) and the results are plotted in Figure 4 for different injection mass cuts. The injection mass cut represents the cut in mass coordinate that separates the material from the star which has been incorporated into the solar nebula to the material which was lost to space or fell back onto the star (Meyer & Clayton 1999). Following Dauphas et al. (2008), we fixed $\Delta t = 0$ and our calculations give lower limits on collateral effects. If the ^{70}Zn was injected by a Type II supernova, the normal (terrestrial) abundance of ^{70}Zn in meteorites at the $\pm 0.4\epsilon$ level limits the possible heterogeneity of ^{60}Fe to less than $\pm 15\%$ dispersion around the average (except for a star of 25 $M_\odot$ and an injection mass cut between 6.7 and 7.1 $M_\odot$ where the allowed dispersion can go up to 50%). Meyer (2005) proposed as the most likely candidate for injection of short-lived radionuclides in the early solar system a 25 $M_\odot$ supernova with an injection mass cut of 5.5 $M_\odot$. In that case, the possible heterogeneity of ^{60}Fe would be less than $\pm 12\%$ dispersion around the average. This confirms previous conclusions from Dauphas et al. (2008) based on ^{64}Ni and ^{58}Fe isotopic measurements. If the source of the ^{60}Fe is an AGB star, Dauphas et al. (2008) showed that the limitation in the variability of ^{60}Fe is even more stringent ($\pm 0.2\%$). This level of homogeneity is also consistent with recent results on ^{26}Al (Thrane et al. 2006; Jacobsen et al. 2008) and with a dynamical model of injection of passive tracers in the protoplanetary disk (Boss 2007). One must however keep in mind that Zn is a moderately volatile element with a 50% condensation temperature of 730 K (Lodders 2003) and fractionation between dust and vapor may have occurred in the circumstellar envelopes (Van Winckel et al. 1992). Thus, Zn behaviors, even if difficult to quantify, might have been decoupled from Fe and its isotopes may have been more thoroughly homogenized than Fe. In that respect, and as was suggested by Dauphas et al. (2008), collateral effects on isotopes from the same element (i.e., ^{58}Fe) provide the most stringent constraints on the distribution of ^{60}Fe, though ^{70}Zn strengthens previous conclusions obtained with ^{58}Fe that ^{60}Fe variability is limited to less than 15% dispersion around the average in the inner part of the disk.

5. CONCLUSION

High-precision data on all five Zn natural isotopes show no resolvable deviation from terrestrial values (after correction of mass-dependent isotopic fractionation). In particular, the neutron-rich isotope ^{70}Zn has terrestrial composition at a precision of $\pm 0.4\epsilon$. Assuming that Fe and Zn have not been decoupled during injection, the lack of variation for ^{70}Zn isotopic abundance constrains the possible heterogeneity of ^{60}Fe to less than 15% dispersion around average. These results agree with the conclusions of Dauphas et al. (2008) and with dynamical modeling (Boss 2007). This homogeneity is valid at the scale of meteorite parent bodies and does not preclude possible mineral scale heterogeneities as has been documented in CAIs (Loss & Lugmair 1989; Völkening & Papanastassiou 1990).

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